

# Exhibit D

Docket No.: 28216/38681B  
(PATENT)

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

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In re Patent Application of:  
Ashoke K. SenGupta et al.

Application No.: 10/626,009

Confirmation No.: 9906

Filed: July 24, 2003

Art Unit: 1713

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For: VISCOUS COMPOSITIONS CONTAINING  
HYDROPHOBIC LIQUIDS

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Examiner: W. K. Cheung

**THIRD DECLARATION OF ASHOKE K. SENGUPTA**  
**UNDER 37 C.F.R. §1.132**

MS Amendment  
Commissioner for Patents  
P.O. Box 1450  
Alexandria, VA 22313-1450

Dear Sir:

NOW COMES ASHOKE K. SENGUPTA, Declarant herein, and states as follows:

1. I am a co-inventor of the invention disclosed and claimed in the above-identified patent application.

2. I am presently employed by AMCOL International Corporation, Arlington Heights, Illinois, assignee of the above-identified patent application. My present title is Senior Principal Scientist. I have been employed by AMCOL since 2000, engaged in research for developing sunscreen additives, polymer-modified clay-based thickeners for nonpolar solvents, deposition-aids for depositing oily/hydrophobic actives from cleansing compositions, and polymeric and clay-based coagulation aids for wastewater.

3. Prior to my employment at AMCOL, I was employed at Borregaard, a Norwegian specialty chemical company, as a Senior Research Scientist, involved in

developing technologies for polymeric dispersants and surface modifiers used in concrete and mining operations, emulsifiers, and coagulation-aids.

4. I earned a B.S. (Honors) in chemical engineering from Jadavpur University, India (1987) and a Ph.D. in chemical engineering from Tulane University, New Orleans, LA (1994), studying colloidal phenomena related to dispersion stability, electrokinetic transport of colloids, polymeric emulsifiers, and stability of dispersions in the presence of polymers.

5. I have conducted research in numerous areas involving dispersion/surface-modification technologies. I have published in the Journal of Colloid and Interface Science and the Journal of the Chemical Society, Faraday Transactions, and have presented at numerous professional and scientific conferences. I am a named inventor in several patents and patent publications.

6. I have read and understand the Office Action dated February 1, 2007, which was issued in connection with U.S. Patent Application Serial No. 10/626,009. I also have read and understand published PCT application WO 01/01949 A1.

7. Organophilic clays (layered silicates), known in the art as thickeners for hydrophobic liquids, traditionally have been produced by surface modifying the clay surface using long chain (C14-22) quaternary ammonium compounds (QAC). For health, odor, and skin irritation-related issues, these QACs, especially those derived from tallow and/or containing aromatic groups, preferably are avoided in formulating personal care and cosmetic products. Both the QACs claimed in the '218 patent contain benzyl groups (aromatic), one of which even is derived from tallow. A critical object of the present invention was to be able to produce organoclay-based thickeners for hydrophobic liquids, without having to use QACs as the clay surface modifier.

8. Accordingly, the present claims are directed to a composition capable of thickening hydrophobic liquids, comprising (a) a smectite clay, having plate-like particles (referred to as platelets), wherein the surfaces of the smectite clay are modified by (b) an amphipathic copolymer. The amphipathic copolymer comprises (i) a first comonomer that when homopolymerized generates a hydrophilic polymer that is insoluble in hydrophobic

liquids and (b) a second comonomer that when homopolymerized generates a hydrophobic polymer that is soluble in a hydrophobic liquid. Unexpectedly, it has been found that this amphipathic copolymer is capable of delaminating/exfoliating (i.e., separating clay platelets across their face-surfaces from stacks of tightly aggregated platelets) clay platelets in a hydrophobic liquid, in a manner such that the delaminated/exfoliated clay platelets then can thicken the hydrophobic liquid medium in which they are dispersed. Normally dispersions tend to exhibit thinning, rather than thickening, when the dispersed particles are surface-treated with polymeric surface modifiers. Accordingly, while thinning is the norm for polymeric surface modifiers, the present invention provides the opposite, i.e., thickening.

9. A wide use of polymers, both homopolymers and copolymers, is as a dispersing agent or dispersant. A polymeric dispersing agent enables suspended particles to repel one another, i.e., to counteract their inherent mutual attraction, by appropriately modifying the particle surface. Such interparticle repulsion, known as steric repulsion, stabilizes the particles against agglomeration or flocculation, and hence the term "steric stabilizer" for these dispersants.

10. With respect to the rheological effects of a dispersing agent, a dispersing agent generally tends to reduce the viscosity of concentrated dispersions by minimizing agglomeration of suspended particles. More particularly, suspended particles are said to be "agglomerated" when they remain so strongly attracted to their surrounding particles that they cannot move independently of these neighboring particles under an applied force, even though they may be physically separated from one another. "Moving independently" means that any given particle is able to move in any direction, regardless of the direction in which its neighboring particles move at any instant of time. In effect, the particle-to-particle attraction translates to particulate structures formed in the dispersion, within which the suspending liquid remains partially or wholly entrapped. As long as an externally applied force (e.g., a shear force) is unable to break apart these particulate structures, and thus release the entrapped suspending liquid, the dispersion remains viscous. The entrapped liquid is no longer available to impart fluidity for the dispersion, and hence the increased viscosity. A dispersant's function is to minimize particle-to-particle "bonds" or attractions that result in particulate structures. In general, the steric stabilizers, namely the polymeric dispersants, are particularly effective in reducing interparticle attraction. Therefore, the expected result of using steric stabilizers is a reduction in dispersion viscosity.

11. To demonstrate the new and unexpected benefits provided by the compositions of the present invention, fourteen dispersion samples, summarized in Table I of paragraph 13, were prepared under my supervision. The procedure followed for preparing the dispersion samples involved the following steps:

- The hydrophobic liquid (octyl stearate) or a solution of PEG-30 dipolyhydroxystearate in octyl stearate was weighted out in a glass beaker.
- The inorganic solid was added in small portions to the hydrophobic liquid or to the polymer solution in the hydrophobic liquid, while mixing the batch in a high-shear, rotor-stator homogenizer (Silverson homogenizer). For a given inorganic solid, the total amount of solid added corresponds to the amount at which the dispersion viscosity was well within the workable range of being able to shear the dispersion in the said batch mixer (it becomes difficult to shear a dispersion in a batch homogenizer if the dispersion viscosity is too high).
- Upon completing the addition of the inorganic solid, the resulting dispersion was sheared in the homogenizer for a period of 5 minutes at a speed of 4,500 - 5,500 rpm.
- The Brookfield viscosity of the dispersion was measured using a Brookfield RVT viscometer operated at 0.5 rpm and 5 rpm of spindle speed at a temperature of 25 - 35°C.

The results of the dispersion tests are given in Table I of paragraph 13, which show that treatment with PEG-30 dipolyhydroxystearate resulted in thinning (and no appreciable thickening) of dispersions of all inorganic solids tested that were disclosed by Lukenbach et al. In contrast, the claimed smectite clay dispersions (Sample No. 2 and 16) showed a relatively high viscosity when PEG-30 dipolyhydroxystearate was used as the clay surface-modifier. In the absence of the polymer (Sample No. 1 and 15), the clay dispersion separated into a layer of the hydrophobic liquid and a sediment layer of settled clay solids, soon after the dispersion was prepared.

12. It is evident from Table I below that Sample No. 2 and 16, utilizing a copolymer of the present invention with a claimed smectite clay, exhibited substantial thickening of the hydrophobic solvent. Note that, absent the copolymer (Sample No. 1 and 15) the clay alone failed to thicken the solvent. Unlike the claimed combination of the copolymer with the claimed smectite clay, the dispersions containing the claimed copolymer together with talc (Sample No. 6); calcium silicate (Sample No. 4); fumed silica (Sample

No. 8); fumed alumina (Sample No. 10); mica (Sample No. 12); and sodium magnesium fluorosilicate (Sample No. 14) did not show any appreciable thickening, but rather thinning in most cases. In particular, sodium magnesium fluorosilicate, a synthetic, fluorine-modified (substituted) layered silicate that bears some compositional resemblance to a claimed smectite clay, hectorite, did not produce any appreciable thickening in the presence of PEG-30 dipolyhydroxystearate, albeit the claimed smectite clay with no fluorine-modification, hectorite (Sample No. 16), showed a considerable thickening under a similar treatment with PEG-30 dipolyhydroxystearate. The slight increase in viscosity in Sample No. 14 (with polymer) as compared to Sample No. 13 (with no polymer) is primarily attributed to the viscosity-gain due to the dissolved polymer in the polymer solution, based on the viscosity results for Sample No. 17 and 18. It is therefore quite understandable why Lukenbach et al. specified the use of sodium magnesium fluorosilicate and the other foregoing solids merely as reflectants and not as thickening agents. Noting the predominant trend (from Table I) - thinning of dispersions in hydrophobic liquids upon treatment with PEG-30 dipolyhydroxystearate, it is virtually impossible to imagine that someone skilled in the art would attempt producing viscous smectite clay dispersions in hydrophobic liquids by surface-treating the clay with the said copolymer. The experimental findings noted herein, therefore, stand as further evidence in support of the unexpected nature of the present invention.

13. Table I

Sample No.	Solid, Weight %	Surface Weight %	Modifier,	Hydrophobic Liquid	Brookfield Viscosity, cps	
					0.5 rpm	5 rpm
1	Bentonite clay, 17.3	None		Octyl Stearate	100	40
2	Bentonite clay, 17.3	PEG-30 Dipolyhydroxystearate, 17.3		Octyl Stearate	>8,000,000	>6,600,000
3	Calcium Silicate, 17.3	None		Octyl Stearate	272,000	11,200
4	Calcium Silicate, 17.3	PEG-30 Dipolyhydroxystearate, 17.3		Octyl Stearate	2,000	500
5	Talc, 20.0	None		Octyl Stearate	2,400	440
6	Talc, 20.0	PEG-30 Dipolyhydroxystearate, 20.0		Octyl Stearate	1,600	240
7	Fumed Silica, 3.8	None		Octyl Stearate	469,000	44,000
8	Fumed Silica, 3.8	PEG-30 Dipolyhydroxystearate,		Octyl Stearate	200	80

		3.8			
9	Fumed Alumina, 17.3	None	Octyl Stearate	28,000	4,320
10	Fumed Alumina, 17.3	PEG-30 Dipolyhydroxystearate, 17.3	Octyl Stearate	200	100
11	Mica, 17.3	None	Octyl Stearate	4,000	640
12	Mica, 17.3	PEG-30 Dipolyhydroxystearate	Octyl Stearate	2,000	240
13	Sodium Magnesium Fluorosilicate, 17.3	None	Octyl Stearate	80	32
14	Sodium Magnesium Fluorosilicate, 17.3	PEG-30 Dipolyhydroxystearate, 17.3	Octyl Stearate	160	60
15	Hectorite Clay, 17.3	None	Octyl Stearate	80	32
16	Hectorite clay, 17.3	PEG-30 Dipolyhydroxystearate, 17.3	Octyl Stearate	280,000	59,200
17	No solid	No polymer	Octyl Stearate	100	22
18	No solid	PEG-30 Dipolyhydroxystearate solution in octyl stearate, mixed in the proportion of 17.3 parts of the polymer dissolved in 65.4 parts of octyl stearate		140	48

14. As noted, an unexpected benefit of the present invention is providing smectite clay-based thickeners for hydrophobic liquids by modifying the clay surface with a polymeric surface modifier, as opposed to a traditional quaternary ammonium compound QAC.

15. All statements made herein of my own knowledge are true and all statements made on information and belief are believed to be true; further, these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001, Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or document or any patent resulting therefrom.

Dated: 2-4-08

Ashoke SenGupta  
Ashoke K. SenGupta